

# Temperature Dependent Magnetic Anisotropy in (Ga,Mn)As Layers

M. Sawicki,<sup>1,\*</sup> F. Matsukura,<sup>1,2</sup> A. Idziaszek,<sup>1</sup> T. Dietl,<sup>1,3</sup>  
G.M. Schott,<sup>4</sup> C. Ruester,<sup>4</sup> C. Gould,<sup>4</sup> G. Karczewski,<sup>1,4</sup> G. Schmidt,<sup>4</sup> and L.W. Molenkamp<sup>4</sup>

<sup>1</sup>*Institute of Physics, Polish Academy of Sciences, al. Lotników 32/46, 02-668 Warszawa, Poland*

<sup>2</sup>*Laboratory for Nanoelectronics and Spintronics,*

*Research Institute of Electrical Communication, Tohoku University,  
Katahira 2-1-1, Aoba-ku, Sendai 980-8577, Japan and*

*ERATO Semiconductor Spintronics Project, Japan Science and Technology Agency, Japan*

<sup>3</sup>*ERATO Semiconductor Spintronics Project, al. Lotników 32/46, PL-02668 Warszawa, Poland  
and Institute of Theoretical Physics, Warsaw University, PL-00681 Warszawa, Poland*

<sup>4</sup>*Physikalisches Institut (EP3), Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany*

(Dated: February 2, 2008)

It is demonstrated by SQUID measurements that (Ga,Mn)As films can exhibit perpendicular easy axis at low temperatures, even under compressive strain, provided that the hole concentration is sufficiently low. In such films, the easy axis assumes a standard in-plane orientation when the temperature is raised towards the Curie temperature or the hole concentration is increased by low temperature annealing. These findings are shown to corroborate quantitatively the predictions of the mean-field Zener model for ferromagnetic semiconductors. The in-plane anisotropy is also examined, and possible mechanisms accounting for its character and magnitude are discussed.

PACS numbers: 75.50.Pp, 75.30.Gw, 75.70.-i

## I. INTRODUCTION

The discovery of carrier-mediated ferromagnetism in (III,Mn)V dilute magnetic semiconductors (DMS) grown by molecular beam epitaxy (MBE) has made it possible to combine complementary properties of semiconductor quantum structures and ferromagnetic systems in single devices, paving the way for the development of functional semiconductor spintronics.<sup>1</sup> Therefore, the understanding of magnetic anisotropy in these systems and the demonstration of methods for its control is timely and important. It has been known, since the pioneering works of Munekata *et al.*<sup>2</sup> and Ohno *et al.*,<sup>3,4</sup> that ferromagnetic (In,Mn)As and (Ga,Mn)As films are characterized by a substantial magnetic anisotropy. Shape anisotropy effects can explain neither the direction nor the large magnitude of the observed anisotropy field  $\mu_0 H_A$  in these dilute magnetic materials.

It has been found by studies of the anomalous Hall effect<sup>3,4</sup> and ferromagnetic resonance<sup>5</sup> that the direction of the easy axis is rather controlled by epitaxial strain in these systems. Generally, for layers under tensile biaxial strain [like (Ga,Mn)As on an (In,Ga)As buffer] perpendicular-to-plane magnetic easy axis has been observed (perpendicular magnetic anisotropy, PMA). In contrast, the layers under compressive biaxial strain [as canonical (Ga,Mn)As on a GaAs substrate] have been found to develop in-plane magnetic easy axis (in-plane magnetic anisotropy, IMA). At first glance this sensitivity to strain appears surprising, as the Mn ions are in the orbital singlet state  $^6A_1$ .<sup>6</sup> For such a case the orbital momentum  $L = 0$ , so that effects stemming from the spin-orbit coupling are expected to be rather weak and, indeed, electron paramagnetic resonance studies of Mn in GaAs have led to relevant spin Hamiltonian param-

eters by two orders of magnitude too small to explain the values of  $\mu_0 H_A$ .<sup>7</sup>

In the system in question, however, the ferromagnetic Mn-Mn exchange interaction is mediated by the band holes, whose Kohn-Luttinger amplitudes are primarily built up of anion p orbitals. Furthermore, in semiconductors, in contrast to metals, the Fermi energy is usually smaller than the atomic spin-orbit energy. Hence, as noted by some of the present authors and co-workers,<sup>8,9</sup> the confinement or strain-induced anisotropy of the valence band can result in a sizable anisotropy of spin properties. Indeed, the quantitative calculation within the mean-field Zener model,<sup>9,10,11</sup> in which the valence band is represented by the  $6 \times 6$  Luttinger Hamiltonian, explains the experimental values of  $H_A$  in (Ga,Mn)As with an accuracy better than a factor of two.<sup>10,12</sup> Moreover, by combining theories of magnetic anisotropy<sup>9,10</sup> and of magnetic stiffness,<sup>13</sup> it has been possible to describe the width of stripe domains in (Ga,Mn)As PMA films.<sup>12</sup> However, the theories in question<sup>10,11</sup> contain a number of predictions that call for a detail experimental verification.

In this paper we present magnetic anisotropy studies carried out by direct magnetization measurements in a dedicated superconducting quantum interference device (SQUID) magnetometer. Our results show that films of (001) (Ga,Mn)As on GaAs with appropriately low values of hole density  $p$  do exhibit PMA and that in some of them a clear temperature-induced reorientation of the easy axis from [001] to  $\langle 100 \rangle$  occurs. This peculiar behavior is actually quite universal and has been observed by some of us and co-workers in the case of (Al,Ga,Mn)As (Ref. 14) and of (Ga,Mn)As obtained under different growth conditions.<sup>15</sup> Importantly, the effect vanishes in samples with higher hole concentrations, which we obtain

by low temperature annealing. By a quantitative comparison of these findings to results of theoretical computations, we demonstrate that the temperature-induced reorientation of the easy axis from the perpendicular (PMA) to the in-plane (IMA) orientation corroborates the theoretical expectations referred to above.<sup>10</sup> At the same time our magnetization data confirm a peculiar character of the *in-plane* anisotropy, which can be inferred from previous transport<sup>16</sup> and magnetooptical<sup>17,18</sup> measurements. We discuss possible reasons and consequences of this specific temperature-dependent in-plane magnetic anisotropy of the (Ga,Mn)As/GaAs material system.

## II. SAMPLES AND EXPERIMENTAL

The (Ga,Mn)As films with a thickness of 400 nm were deposited by MBE onto (001) GaAs substrates at 220°C under an As<sub>4</sub>/Ga beam equivalent pressure ratio of 5, as described previously.<sup>19</sup> High resolution x-ray diffraction shows good crystal quality (Ga,Mn)As with the rocking curve widths comparable to those of the GaAs substrate and pronounced finite-thickness fringes, indicating flat interfaces and surfaces. The layers are pseudomorphic with respect to the GaAs substrate, as verified by reciprocal space maps around the asymmetric [115] reflection. We present results for samples with Mn concentration  $x = 5.3\%$  and  $3\%$ , and biaxial strain  $\varepsilon_{xx} = -0.27\%$  and  $-0.16\%$ , respectively, as established by x-ray diffraction measurements.<sup>19</sup> In order to trace the evolution of magnetic anisotropy with the hole concentration, the  $x = 5.3\%$  sample was divided into six pieces, five of them being annealed at 165°C in air for different times between 28 and 200 h, which led to an increase of  $T_C$  up to a factor of two. This sensitivity to annealing reflects the presence of self-compensation by mobile donor defects, which are neutralized at the surface.<sup>20</sup>

Magnetization measurements are carried out in the SQUID magnetometer down to 5 K. By design, the signal detection axis is aligned with the direction of the magnetic field (vertically). As a result the system is sensitive only to the vertical component of the magnetization vector. Special care is put forward to screen the sample from external magnetic fields and to keep the parasite remanent fields generated by the magnet at the lowest possible level. The Meissner effect of a pure lead sample confirms that even after an excursion to the field of 1 kOe, the remanent field remains below 100 mOe. Such a low value is essential for successful magnetic remanence studies or for measurements at temperatures close to  $T_C$ , where the coercivity of (Ga,Mn)As drops into a sub-Oersted regime. For studies of in-plane anisotropy, (Ga,Mn)As layers were initially shaped by chemical etching into circles of approximately 5 mm in diameter. However, detailed studies shows that this is an unnecessary precaution, since even rectangular 2:1 shapes have the same properties as circled samples.

## III. ORIGIN OF MAGNETIC ANISOTROPY IN FERROMAGNETIC ZINC-BLENDE DMS

The magnetic dipolar anisotropy, or shape anisotropy, is mediated by dipolar interaction. Since it is long range, its contribution depends on the shape of the sample and in thin films the shape anisotropy often results in the in-plane alignment of the moments. The experimental evidence presented in this paper unambiguously proves that perpendicular orientation of spontaneous magnetization is realized in *some* of the investigated (Ga,Mn)As/GaAs layers. Importantly, similar studies<sup>2</sup> showed that PMA is seen in *most* (III,Mn)V layer grown such that they are under tensile biaxial strain. As all are very thin layers (typically a fraction of  $\mu\text{m}$  thick) and of macroscopic lateral dimensions, such an experimental finding points to the existence of a very strong microscopic mechanism that counteracts the shape imposed in-plane arrangement of the magnetization.

Undoubtedly, the sign of the biaxial strain is one of the factors that plays a role in determining of the direction of the magnetic anisotropy. Indeed, at sufficiently high hole concentrations, this is often the dominant factor. However, our experimental results clearly show that in general, additional factor such as hole concentration and temperature also play a role, and that the anisotropy is determined by a combination of these factors. In fact, we will argue that sensitivity to epitaxial strain, hole density and temperature is an ubiquitous property of carrier mediated ferromagnetism and is solely due to the anisotropy of the carrier-mediated exchange interaction reflecting the anisotropic properties of the top of the valence band. This should not be too surprising given that we are dealing with magnetically diluted systems, and that the shape anisotropy fields that must be overcome are not particularly strong. In our case, as for thin films, the shape anisotropy energy per unit volume is given by:  $E = \frac{1}{2}\mu_0 M_S^2 \cos^2 \theta$  ( $M_S$  is the saturation magnetization and  $\theta$  is the angle that  $M_S$  subtends to the plane normal), which gives the anisotropy field  $\mu_0 H_A = \mu_0 M_S$  only of about 0.06 T for 5 % (Ga,Mn)As, as compared to 2.2 T for iron.

Knowing the p-d exchange energy and  $k \cdot p$  parameters of the valence band it is possible to compute magnetic anisotropy energy in the studied compounds.<sup>9,10,11</sup> In fact the published results agree with the experimental data with remarkable good accuracy.<sup>5,21,22,23</sup> Nevertheless, it is instructive to consider a simplified case of the model that is the nearly empty top of the valence band in biaxial strained zinc-blende compounds. When the strain is present the valence band splits and the energetic distance between the heavy-hole  $j_z = \pm 3/2$  and light-hole  $j_z = \pm 1/2$  subbands depends on strain, see Fig. 1. For the biaxial compressive strain the ground state subband assumes a heavy-hole character. Then, if only the ground state subband is occupied, the hole spins are oriented along the growth direction. Now, since the p-d exchange interaction has a scalar form,  $H_{pd} \sim s \cdot S$ ,

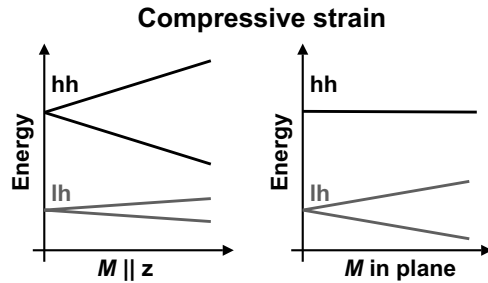


FIG. 1: Scheme of valence band splitting in tetrahedrally coordinated semiconductors for compressive strain and for two orientations of magnetization  $M$  with respect to sample plane.

the in-plane Mn spin magnetization  $M$  will not affect the heavy-hole subband. This means that perpendicular magnetic anisotropy is expected, since only for such magnetization orientation can the holes lower their energy by the coupling to the Mn spins. In the opposite, tensile strain case, the in-plane component of the hole spin is greater than the perpendicular component, so a stronger exchange splitting occurs for the in-plane orientation of  $M$ . Hence, if only the light-hole subband is occupied – the in-plane anisotropy is expected. It is worth remarking here, that PMA is not a unique property of (III,Mn)V ferro-DMS. PMA is typically realized in compressive strained (Cd,Mn)Te QW<sup>8,21</sup> and the recent work showed that in (II,Mn)VI/II,VI structures the magnetic anisotropy can be tailored by adequate strain engineering.<sup>24</sup> For the compressive strain a ferromagnetic state related splitting of the luminescence line is found only for the perpendicular orientation. However, when large enough tensile strain was built into that system the in-plane direction of the easy axis has been observed.

By nature (III,Mn)V DMS systems are heavily populated with holes. But even when the Fermi energy is comparable or larger than the heavy hole - light hole splitting, that is when the strong mixing takes place, we can still talk about either heavy- or light hole like character of the holes. Therefore the lines of reasoning sketched above remain valid to a large extend in (III,Mn)V and such a simplified approach can still serve as a helpful guideline. So, we are going to use it again to sketch how in (III,Mn)V ferro-DMS the direction of the magnetic easy axis can be set or altered by changing hole density and/or temperature.

#### IV. REORIENTATION OF MAGNETIC EASY AXIS

The Zener model dictates that Mn moments, or more accurately their collective macroscopic magnetization, adjusts its orientation to minimize the total energy of the carriers required to support ferromagnetic ordering of the Mn ions. In particular, depending on the Fermi level position within the valence band and/or the value of the exchange splitting (that is depending on magnetization and thus also on temperature) different orientations of magnetization can be required to drive the system to its energy minimum. Therefore, by changing hole concentration or temperature, the corresponding changes of the overall orbital momentum of the hole liquid may force a *spontaneous* reorientation of magnetization. It is relatively easy to trace such an effect when we consider hole-concentration-induced reorientation. Using again Fig. 1 we note that by introducing more holes into the system we populate the second, light-hole, subband for which the ferromagnetic state can only be realized with the in-plane magnetization orientation. This effect gets even stronger since the heavy holes acquire a light hole character on increasing Fermi energy  $E_F$ , and it is therefore expected that at some 'critical' value of the hole concentration, it will be more favourable for the holes to experience in-plane Mn magnetization. So, an isothermal change of the hole density<sup>25</sup> can lead to the out-of-plane to in-plane reorientation of the magnetic anisotropy, providing the population of light-holes gets large enough. A similar mechanism operates when the temperature is used as a handle for the easy axis switching. The only difference is that this time the hh/lh population is changed via temperature induced changes of the valence subbands exchange splitting. Now, since the spin-splitting is proportional to Mn magnetization  $M_s(T)$  that varies according to the Brillouin-like function, the character of magnetic anisotropy depends on the temperature. Accordingly, in compressively strained structures PMA occurs at low both temperatures and hole concentrations, while otherwise IMA will be realized. This implies that there exists a class of samples for which the material parameters are such that within the experimental temperature range a reorientation of the easy axis from easy z-axis to easy plane should occur on increasing temperature. An obvious question arises then about the expected magnitude of changes of hole concentration and temperature required for the reorientation to take place. At this point, however, we want to strongly underline that despite our general understanding of the physical mechanisms responsible for the studied effects, the strong mixing of the valence band states results in such a large anisotropic and non-parabolic valence subbands dispersions that does not allow us to specify a single hh/lh ratio and/or hole density/magnetization/temperature values necessary to trigger magnetization easy axis switch. This has to be computed up to the fullest possible extent of the mean field model and for each and particular sample individually.

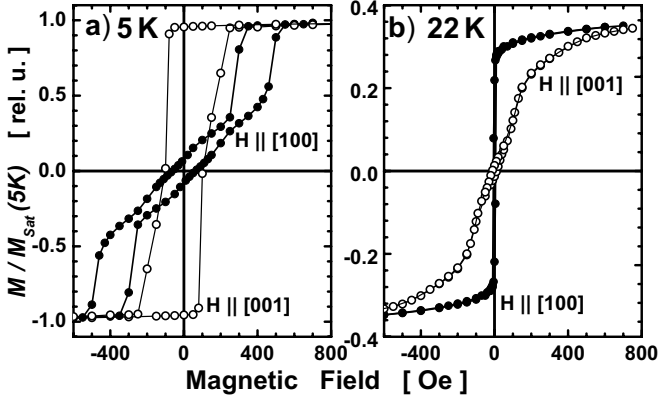


FIG. 2: Field dependence of magnetization normalized by its saturation value at 5 K for two orientations of the film in respect to the magnetic field at two temperatures (a,b) for as grown  $\text{Ga}_{0.947}\text{Mn}_{0.053}\text{As}$ . Full and empty dots denote the data taken for the magnetic field along the [100] and [001] crystal direction, respectively. Note the flip of the easy axis direction from the perpendicular to the in-plane orientation on increasing temperature.

## V. RESULTS AND DISCUSSION

Figures 2(a) and 2(b) present typical hysteresis loops for the non-annealed (Ga,Mn)As sample which exhibits the temperature driven change from PMA to IMA. First, let us note that the sample does exhibit a perpendicular easy axis at low temperatures. As Fig. 2(a) presents, a perfect square hysteresis is obtained when the magnetic field is perpendicular to the film surface and an elongated loop is seen when the in-plane orientation is probed. Remarkably, a reverse behavior is observed *for the same sample* at higher temperatures (Fig. 2b). These findings demonstrate that the easy axis flips from the perpendicular to in-plane orientation on increasing temperature.

In order to trace the reorientation of spontaneous magnetization  $M_s$  more closely, we have examined the temperature dependence of remanent magnetization  $M_{\text{REM}}$ , measured along selected crystallographic directions according to the procedures described in the caption to Fig. 3. As evidenced in the figure we find  $M_{\text{REM}}$  at low temperatures to be tenfold larger for the perpendicular experimental arrangement,  $H_{\text{FC}} \parallel [001]$ , compared to the case of the parallel one,  $H_{\text{FC}} \parallel [100]$ . This reconfirms the appearance of PMA at low temperatures in this film despite the presence of a sizable compressive strain. Large values of perpendicular  $M_{\text{REM}}$  hold until a certain temperature ( $\cong 10$  K in this case) at which a sudden drop of the signal is detected. This is followed by a slow decay, which we attribute to a material inhomogeneity. Further warming does not change this situation, perpendicular  $M_{\text{REM}}$  stays vanishingly small until  $T_C$ . Clearly,  $M_s$  rotates out of [001] direction, and locks itself in the plane of the layer somewhere within this temperature range. We name the temperature at which this rotation takes place the reorientation temperature  $T_R$ . However, the vanish-

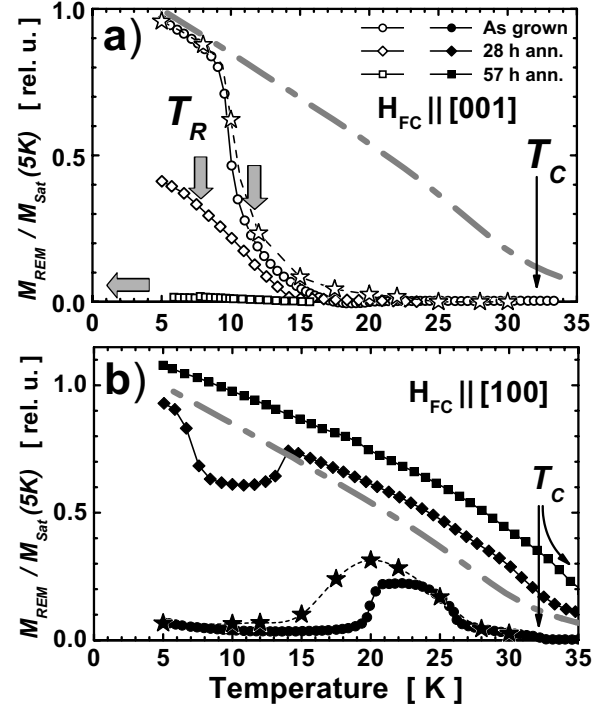


FIG. 3: Temperature dependence of the remanent magnetization as measured in perpendicular [001] (a) and in plane [100] (b) configuration for the  $\text{Ga}_{0.947}\text{Mn}_{0.053}\text{As}$  sample prior to annealing (circles) and after annealing (diamonds and squares). Two experimental methods are presented. Temperature dependent  $M_{\text{REM}}$  (circles), the sample is cooled down through  $T_C$  in the field  $H_{\text{FC}} = 1$  kOe, which is at least by a factor of ten higher than the coercive field  $H_C$ . Then, the field is removed at 5 K, and the measurement of the magnetization component  $M_{\text{REM}}$  along the direction of  $H_{\text{FC}}$  commences on increasing temperature in the residual field  $H_r \leq 100$  mOe. Alternatively, (Isothermal  $M_{\text{REM}}$ , stars) the same  $M_s$  component at selected temperatures  $T < T_C$  is obtained by field cooling from above  $T_C$  to  $T$  and then removing the external magnetic field. Both methods are seen to give essentially the same results. Note that upon annealing the development of the in-plane component of  $M_{\text{REM}}$  is accompanied by an equivalent quench of the perpendicular one. Bulk arrows mark the reorientation temperature  $T_R$  when the cross-over to in-plane magnetic anisotropy takes place. The thick dashed grey lines mark result of FC measurements at 1 kOe for the as grown layer, and thus mimic the temperature dependence of the saturation magnetization.

ing of perpendicular  $M_{\text{REM}}$  is not accompanied by an immediate development of parallel  $M_{\text{REM}}$ . This does not contradict our understanding, since due to equivalence of all in-plane easy cubic directions, the flip of magnetization into the plane results in a *demagnetized* state, characterized by the presence of closure domains, and thus by small overall sample  $M_{\text{REM}}$ . Actually, a sizable increase of the parallel  $M_{\text{REM}}$  between  $T_R$  and  $T_C$  reflects only the presence of the residual field  $H_r$  generated in our magnet. This small field, by breaking up the closure domains, starts to magnetize the film at temperatures when



the domain wall propagation mechanisms get effective. However, since  $H_r$  assumes extremely small value in our system, the sample gets only partially magnetized, which accounts for the differences observed at this temperature range between temperature-dependent REM and isothermal REM measurements, as seen in Fig. 3(b).

As already noted, the temperature-induced cross-over from PMA to IMA described above has been found in other samples of (Al,Ga,Mn)As and (Ga,Mn)As.<sup>14,15</sup> Remarkably, the opposite behavior occurs in (In,Mn)As under a tensile strain, where the easy axis flips from in-plane to out of plane on warming.<sup>26,27</sup> As sketched in the previous section, this stems from the fact that the direction of orbital momentum, which controls magnetic anisotropy, differs for the heavy and light holes, whose relative concentration depends not only on the sign of biaxial strain but on the magnitude of the spin splitting as well. Importantly, striking temperature dependent anisotropy in these systems can, in fact, be inferred from numerical computations presented in the previous theoretical work.<sup>10</sup> In particular, according to Fig. 10 of Ref. 10, there is a range of the hole concentrations for which the easy axis of compressed films is expected to switch from the perpendicular to the in-plane direction on decreasing the valence band spin-splitting (that is on increasing temperature, while the switching in the opposite sense is predicted for the tensile strain).

Figure 4 presents the computed reorientation temperature  $T_R$  as a function of the hole concentration  $p$  for the two studied samples. The theoretical model and material parameters described in detail previously<sup>10</sup> are adopted to obtain this phase diagram. Shape anisotropy is taken into account, and it shifts the PMA  $\rightarrow$  IMA phase boundary by about 20 % towards lower  $p$  values. The same theory is employed to determine the magnitude of  $p$  from the experimental values of  $x$  and  $T_C$ . We see that the theoretical model confirms the appearance of PMA and describes correctly temperature driven phase transformation PMA  $\rightarrow$  IMA in the case of the  $x = 5.3\%$  sample prior to annealing. According to theory, such a phenomenon should vanish for higher values of  $p$ . Indeed, as seen from Fig. 3, if the annealing time and thus the magnitude of  $T_C$  and  $p$  are sufficiently large, these samples exhibit only IMA.

At this point it is worth to emphasize that although the reorientation transition is the general feature of heavily doped ferro-DMS, it is only a sample specific property. In particular, for a given strain if the hole concentration is either too small or too large no reorientation transition is expected for any value of magnetization (temperature). In fact, as Fig. 4 demonstrates, the range of hole densities for which the reorientation can occur is quite narrow. On the other hand, for an appropriate combination of strain and hole concentration, even a minute change of magnetization (temperature) switches the easy axis between the two directions. This feature is confirmed by a comparison of our  $M_{\text{REM}}(T)$  and quasi- $M_s(T)$  data for the as-grown sample. As seen in Fig. 3,  $M_s$  is undoubtedly a smooth and slowly varying function of temperature, and

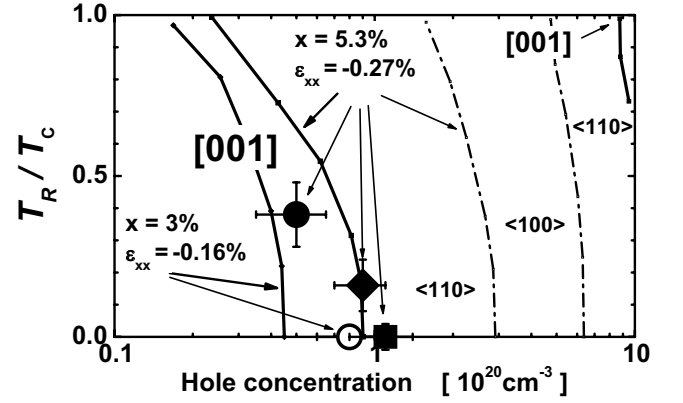


FIG. 4: Experimental (full points, taken from Fig. 3 for  $x = 5.3\%$  sample) and computed values (thick lines) for the ratio of reorientation and Curie temperatures in  $\text{Ga}_{1-x}\text{Mn}_x\text{As}$  for perpendicular to in-plane magnetic anisotropy transition. For  $x = 3\%$  sample (the open circle) this transition is not detected above 5 K, in an agreement with the presented calculations. Dashed lines mark expected temperatures for the in-plane reorientation of the easy axis between  $\langle 100 \rangle$  and  $\langle 110 \rangle$  directions.

despite of this the reorientation does take place.

According to the discussion above, the easy axis assumes the in-plane orientation for typical carrier concentrations in (Ga,Mn)As/GaAs. In this case, according to the theoretical predictions presented in Fig. 4 as well in Fig. 9 of Ref. 10 and in Fig. 6 of Ref. 11, the four-fold magnetic symmetry with the easy axis is expected to switch between  $\langle 100 \rangle$  and  $\langle 110 \rangle$  in-plane cubic axes as a function of  $p$  or  $T$ . This biaxial magnetic symmetry is indeed observed at low temperatures, however with the easy axis assuming exclusively  $\langle 100 \rangle$  in-plane orientation, as observed by us and others.<sup>5,16,17,18,22,28</sup> To our best knowledge, no  $\langle 100 \rangle \leftrightarrow \langle 110 \rangle$  transition has been detected to date. This may indicate that the anisotropy of the hole magnetic moment, neglected in the theoretical calculations,<sup>10,11</sup> stabilizes the  $\langle 100 \rangle$  orientation of the easy axis. It is also possible that the stabilization energy comes from broken magnetic bonds at the film surfaces, an effect put into evidence in the case of (Fe,Co)/GaAs films.<sup>29</sup> However, whether any of these models explains simultaneously the reported recently  $\langle 110 \rangle$  biaxial symmetry in (In,Mn)As/(In,Al)As films,<sup>30</sup> remains to be shown.

Our magnetization data reveal also the existence of a uniaxial anisotropy in films of (Ga,Mn)As/GaAs. As shown in Fig. 4,  $M_{\text{REM}}$  measured along the  $\bar{1}10$  direction vanishes completely above 15 K indicating that this is the hard direction in this film. We also note that when  $M_{\text{REM}}^{[110]}$  vanishes the  $M_{\text{REM}}^{[100]}/M_{\text{REM}}^{[110]}$  ratio drops to  $\sqrt{2}/2$ , the value expected for the easy axis along  $[110]$ . Since the cubic-like anisotropy energy is proportional to  $M_s^4$  whereas the uniaxial one to  $M_s^2$ , the latter dominates at high temperatures, where  $M_s$  is small. We note that, due to the biaxial strain, the initial  $T_d$  point symmetry

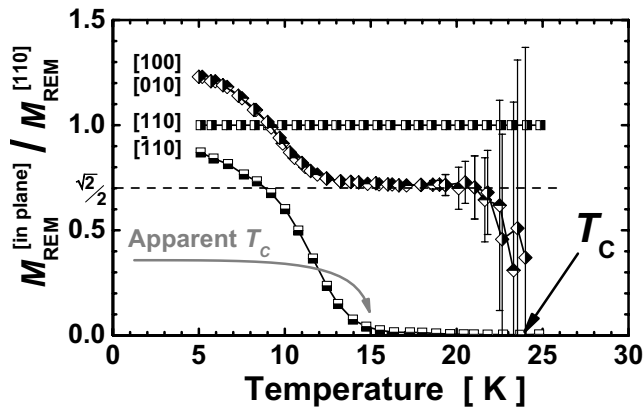


FIG. 5: Experimental evidence for the uniaxial anisotropy along  $[110]$  direction in  $\text{Ga}_{0.97}\text{Mn}_{0.03}\text{As}$  film. The magnetic remanence is measured for four major in-plane directions and its magnitude is normalized by the data of the  $[110]$  case. Note, that the sudden drop of  $M_{\text{REM}}$  along  $[\bar{1}10]$  at  $T \ll T_C$  may wrongly indicate too low value of  $T_C$ , if only this orientation is probed.

of zinc blende structure is lowered to  $D_{2d}$ , for which *no* uniaxial IMA is expected. Furthermore, the magnitude of the corresponding anisotropy field appears to be independent of the film thickness, for both as large as  $7\text{ }\mu\text{m}$ <sup>28</sup> and as low as  $25\text{ nm}$ <sup>31</sup>, what, in particular, rules out the effect of Mn oxide accumulated at the free surface.<sup>20,32</sup> Recently the Argonne - Notre Dame team<sup>18,28</sup> has advocated for an effect connected with surface reconstruction induced preferential Mn incorporation occurring at every step of layer-by-layer growth. At the same time we point out that a unidirectional character of the growth process<sup>18</sup> and/or differences between  $(\text{Ga},\text{Mn})\text{As}/\text{GaAs}$  and  $(\text{Ga},\text{Mn})\text{As}/\text{vacuum}$  interfaces may lower symmetry to  $C_{2v}$  where the three principal directions are:  $[001]$ ,  $[110]$  and  $[\bar{1}10]$ . Since in  $C_{2v}$  they are not equivalent, the  $[110] \Leftrightarrow [\bar{1}10]$  symmetry gets broken, conforming with the

presented results.

In summary, our studies have demonstrated the rich characteristics of magnetic anisotropies in  $(\text{Ga},\text{Mn})\text{As}/\text{GaAs}$ , which—in addition to epitaxial strain—vary with the hole and Mn concentrations as well as with the temperature. According to theory,<sup>9,10,11</sup> these reflect spin anisotropy of the valence band subbands whose shape varies with strain, while the splitting and population depend on magnetization and hole concentration. In particular, the temperature-driven reorientation of the easy axis reflects the equipartition of the valence subband population at  $T$  approaching  $T_C$  and, therefore, reconfirms the crucial role of the valence band holes in the ferromagnetism of  $(\text{III},\text{Mn})\text{V}$  systems. At the same time, our findings have provided the magnetic corroboration for the existence of uniaxial in-plane anisotropy. By group theoretical considerations this unexpected anisotropy can be linked to the top/bottom symmetry breaking, an issue calling for a microscopic modelling. Furthermore, the temperature-induced switching of the easy axis direction revealed by our findings explains the origin of non-standard temperature dependencies of measured magnetization, and indicates that the meaningful determination of  $T_C$  requires measurements for various crystal orientations. We note also that since the direction of spontaneous magnetization depends on the hole concentration which, in turn, can be varied by the electric field<sup>25</sup> or light irradiation,<sup>27</sup> it appears possible to reverse the magnetization in a field-effect transistor having a ferromagnetic semiconductor channel.

### Acknowledgments

The authors thank J. Ferré, H. Ohno, and W. Van Roy for valuable discussions. Support of the FENIKS project (EC:G5RD-CT-2001-0535) is gratefully acknowledged.

\* Electronic address: mikes@ifpan.edu.pl

- <sup>1</sup> For recent reviews, see, H. Ohno, F. Matsukura, and Y. Ohno, JSAP International, No. 5, January 2002, pp. 4-13; S. Wolf, D.D. Awschalom, R.A. Buhrman, J.M. Daughton, S. von Molnr, M.L. Roukes, A.Y. Chtchelkanova, D.M. Treger, *Science* **294**, 1488 (2001); T. Dietl, *Acta Phys. Polon. A* **100** (suppl.), 139 (2001).
- <sup>2</sup> H. Munekata, A. Zaslavsky, P. Fumagalli, and R.J. Gambino, *Appl. Phys. Lett.* **63**, 2929 (1993).
- <sup>3</sup> H. Ohno, F. Matsukura, A. Shen, Y. Sugawara, A. Oiwa, A. Endo, S. Katsumoto, and Y. Iye, in: *Proceedings of the 23rd International Conference on Physics of Semiconductors*, Berlin 1996, edited by M. Scheffler and R. Zimmermann (World Scientific, Singapore, 1996) p. 405.
- <sup>4</sup> A. Shen, H. Ohno, F. Matsukura, Y. Sugawara, N. Akiba, T. Kuroiwa, A. Oiwa, A. Endo, S. Katsumoto and Y. Iye, *J. Cryst. Growth* **175/176**, 1069 (1997).
- <sup>5</sup> X. Liu, Y. Sasaki, and J.K. Furdyna, *Phys. Rev. B* **67**, 205204 (2003).

- <sup>6</sup> J. Szczytko, A. Twardowski, K. Swiatek, M. Palczewska, M. Tanaka, T. Hayashi, and K. Ando, *Phys. Rev. B* **60**, 8304 (1999).
- <sup>7</sup> O.M. Fedorych, E.M. Hankiewicz, Z. Wilamowski, and J. Sadowski, *Phys. Rev. B* **66**, 045201 (2002).
- <sup>8</sup> T. Dietl, A. Haury, Y. Merle d'Aubigné, *Phys. Rev. B* **55**, R3347 (1997).
- <sup>9</sup> T. Dietl, H. Ohno, F. Matsukura, F. Cibert and D. Ferand, *Science* **287**, 1019 (2000).
- <sup>10</sup> T. Dietl, H. Ohno, and F. Matsukura, *Phys. Rev. B* **63**, 195205 (2001).
- <sup>11</sup> M. Abolfath, T. Jungwirth, J. Brum, and A.H. MacDonald, *Phys. Rev. B* **63**, 054418 (2001).
- <sup>12</sup> T. Dietl, J. König, and A.H. MacDonald, *Phys. Rev. B* **64**, 241201(R) (2001).
- <sup>13</sup> J. König, T. Jungwirth, and A.H. MacDonald, *Phys. Rev. B* **64**, 184423 (2001).

- <sup>14</sup> K. Takamura, F. Matsukura, D. Chiba, and H. Ohno, Appl. Phys. Lett. **81**, 2590 (2002).
- <sup>15</sup> M. Sawicki, F. Matsukura, T. Dietl, G. M. Schott, C. Rueter, G. Schmidt, L. W. Molenkamp and G. Karczewski, Superconductivity/Novel Magnetism **16**, 7 (2003).
- <sup>16</sup> S. Katsumoto, A. Oiwa, Y. Iye, H. Ohno, F. Matsukura, A. Shen and Y. Sugawara, phys. status solidi (b) **205**, 115 (1998); H.X. Tang, R.K. Kawakami, D.D. Awschalom and M.L. Roukes, Phys. Rev. Lett. **90**, 107201 (2003); K.Y. Wang, K.W. Edmonds, R.P. Campion, L.X. Zhao, A.C. Neumann, C.T. Foxon, B.L. Gallagher, P.C. Main, Presented at ICPS-26, July 2002, cond-mat/0211697.
- <sup>17</sup> D. Hrabovsky, E. Vanelle, A. R. Fert, D. S. Yee, J. P. Redoules, J. Sadowski, J. Kanski, and L. Ilver, Appl. Phys. Lett. **81**, 2806 (2002).
- <sup>18</sup> U. Welp, V.K. Vlasko-Vlasov, X. Liu, J.K. Furdyna and T. Wojtowicz, Phys. Rev. Lett. **90**, 167206 (2003).
- <sup>19</sup> G.M. Schott, W. Faschinger, and L.W. Molenkamp, Appl. Phys. Lett. **79**, 1807 (2001).
- <sup>20</sup> K.W. Edmonds, P. Boguslawski, K.Y. Wang, R.P. Campion, S.N. Novikov, N.R.S. Farley, B.L. Gallagher, C.T. Foxon, M. Sawicki, T. Dietl, M.B. Nardelli and J. Bernholc, Phys. Rev. Lett. **92**, 037201 (2004).
- <sup>21</sup> H. Boukari, P. Kossacki, M. Bertolini, D. Ferrand, J. Cibert, S. Tatarenko, A. Wasiela, J. A. Gaj, and T. Dietl, Phys. Rev. Lett. **88**, 207204 (2002).
- <sup>22</sup> G.P. Moore, J. Ferr, A. Mougin, A. Moreno and L. Dweritz, J. Appl. Phys. **94**, 4530 (2003).
- <sup>23</sup> P. Van Dorpe, Z. Liu, W. Van Roy, V.F. Motsnyi, M. Sawicki, G. Borghs, and J. De Boeck, Appl. Phys. Lett. **84** (2004) 3495.
- <sup>24</sup> P. Kossacki, W. Pacuski, W. Małana, J. A. Gaj, M. Bertolini, D. Ferrand, S. Tatarenko and J. Cibert, Physica E **21**, 943 (2004).
- <sup>25</sup> H. Ohno, D. Chiba, F. Matsukura, T. Omiya, E. Abe, T. Dietl, Y. Ohno, and K. Ohtani, Nature **408**, 944 (2000).
- <sup>26</sup> T. Endo, T. Ślupski, S. Yanagi, A. Oiwa, and H. Munekata, unpublished.
- <sup>27</sup> X. Liu, W.L. Lim, L.V. Titova, T. Wojtowicz, M. Kutrowski, K.J. Yee, M. Dobrowolska, J.K. Furdyna, S.J. Potashnik, M.B. Stone, P. Schiffer, I. Vurgaftman, J.R. Meyer, Physica E **20**, 370 (2004).
- <sup>28</sup> U. Welp, V.K. Vlasko-Vlasov, X. Liu, J.K. Furdyna, and T. Wojtowicz, Appl. Phys. Lett. **85**, 260 (2004).
- <sup>29</sup> M. Dumm, B. Uhl, M. Zöfl, W. Kipferl, and G. Bayreuther, J. Appl. Phys. **91**, 8763 (2002).
- <sup>30</sup> X. Liu, W-L Lim, Z. Ge, S. Shen, M. Dobrowolska, J. K. Furdyna and T. Wojtowicz, *27th International Conference on Physics of Semiconductors*, Flagstaff AZ, USA, July 2004, abstract, unpublished (2004).
- <sup>31</sup> M. Sawicki, K-Y Wang, K.W. Edmonds, R.P. Campion, C.R. Staddon, N.R.S. Farley, C.T. Foxon, E. Papis, E. Kamiska, A. Piotrowska, T. Dietl and B.L. Gallagher, in preparation (2004).
- <sup>32</sup> J.K. Furdyna, T. Wojtowicz, X. Liu, K. M. Yu and W. Walukiewicz, J. Phys.: Condens. Matter, in press (2004).